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TUNABLE OPTICAL SOURCES

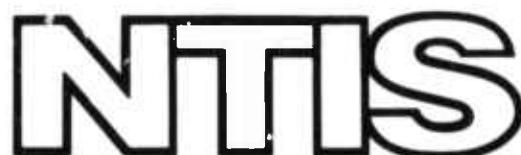
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FINAL REPORT

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M. L. Report No. 2338

August 1974



Microwave Laboratory  
W. W. Hansen Laboratories of Physics  
Stanford University  
Stanford, California

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Scientific Personnel

on

U. S. Army Research Office (Durham)

Contract No. DA-ARO-D-31-124-72-G-184

Final Report

S. E. Harris	Principal Investigator
R. L. Byer	Associate Professor
J. F. Young	Research Associate
R. L. Herbst	Research Associate
D. M. Bloom	Research Assistant
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## INTRODUCTION

Work on this grant has been concerned with new techniques for the development of sources of tunable radiation and on techniques for spectral up-conversion of both incoherent and coherent infrared sources. Emphasis has been placed on the infrared region of the spectrum. Substantial progress has been made in the general areas of nonlinear optical materials, optical parametric oscillators, and nonlinear optical techniques in metal vapors and gases. In the following pages we give abstracts of all publications which have appeared or will appear under this grant.

**Efficient Parametric Mixing in CdSe<sup>†</sup>**

R. L. Herbst and R. L. Byer

Microwave Laboratory, Stanford University, Stanford, California 94305  
(Received 25 August 1971)

We have obtained the first phase-matched nonlinear interaction in CdSe and have observed a 35% conversion efficiency for mixing  $10.6 \mu$  with a pump at  $1.833 \mu$  to generate a signal at  $2.2 \mu$ . The mixing process phase matches at  $77^\circ$  to the optic axis and confirms the predicted phase-matching angle. The measured nonlinear coefficient value of  $2.5 \times 10^{-22}$  mks agrees with previous results. The mixing experiment shows that an angle-tuned or pump-tuned infrared parametric oscillator is possible using CdSe as the nonlinear element.

## Singly resonant CdSe infrared parametric oscillator\*

R.L. Herbst and R.L. Byer

*Microwave Laboratory, Stanford University, Stanford, California 94305*

(Received 15 May 1972)

We have demonstrated an infrared singly resonant parametric oscillator using CdSe as the nonlinear crystal. The oscillator operates with either a resonant signal near  $2.2\text{ }\mu$  or resonant idler in the  $9.8\text{- to }10.4\text{-}\mu$  region. Using a Q-switched Nd: YAG laser operating at  $1.833\text{ }\mu$  as a pump source, we have observed thresholds of 550 W and up to 40% conversion efficiency. The angle-tuned oscillator operates at room temperature with a  $2\text{-cm}^{-1}$  bandwidth.

## **Generation of 1182-Å radiation in phase-matched mixtures of inert gases\***

**A.H. Kung, J.F. Young, and S.E. Harris**

*Microwave Laboratory, Stanford University, Stanford, California 94305*  
(Received 7 December 1972)

Coherent radiation at 1182 Å is obtained by third-harmonic generation in a phase-matched mixture of Xe and Ar. For generation from 3547 to 1182 Å, Xe is negatively dispersive, and phase matching is obtained at a ratio of Xe:Ar=1:430. A conversion efficiency of 2.8% is obtained at an input power of 13 MW. As predicted by theory the conversion efficiency increases linearly to the limit of our available input power.

## OPTICAL THIRD HARMONIC GENERATION

### IN ALKALI METAL VAPORS\*

R. B. Miles<sup>†</sup> and S. E. Harris  
Microwave Laboratory  
Stanford University  
Stanford, California 94305

#### ABSTRACT

The paper considers third harmonic generation in phase matched mixtures of alkali metal vapors and inert gases. Calculations show that the combination of near resonant nonlinear susceptibilities, the ability to phase match, and the relatively high UV transparency of these vapors, should allow high conversion efficiency for picosecond laser pulses with a peak power of  $10^8 - 10^9$  watts. Calculations of the nonlinear susceptibility and of the ratio of xenon atoms to metal vapor atoms which is necessary to achieve phase matching are given for each of the alkalies as a function of incident laser wavelength. Processes which limit the allowable peak power density and energy density are discussed, and guides for determining the metal vapor pressure, cell length, and beam area are given.

## Generation of Vacuum-Ultraviolet and Soft-X-Ray Radiation Using High-Order Nonlinear Optical Polarizabilities\*

S. E. Harris

*Microwave Laboratory, Stanford University, Stanford, California 94305*

(Received 30 May 1973)

The harmonic or sum-frequency power generated in the last coherence length of a low-density atomic species is calculated subject to the condition that the applied electric field be bounded by the multiphoton absorption or ionization limit. It is shown that higher-order polarizations may equal or exceed lower-order polarizations. Calculations are given for generation at 1773 and 1064 Å in Xe, and at 236, 169, and 177 Å in Li<sup>+</sup>.

## **Stimulated emission in multiple-photon-pumped xenon and argon excimers**

**S. E. Harris, A. H. Kung, E. A. Stappaerts, and J. F. Young**

*Microwave Laboratory, Stanford University, Stanford, California 94305*  
(Received 14 May 1973)

Multiple photon absorption of laser radiation at 2660 and 3547 Å is used to pump excimers of Xe and Ar. Line narrowing is observed in bands  $\sim 100$  Å wide, centered at 1730 and 1260 Å, respectively.

GENERATION OF ULTRAVIOLET AND VACUUM ULTRAVIOLET RADIATION\*

by

S. E. Harris, J. F. Young, A. H. Kung, D. M. Bloom, and G. C. Bjorklund

Microwave Laboratory  
Stanford University  
Stanford, California

ABSTRACT

The paper describes the use of nonlinear optical techniques for the generation of coherent radiation at ultraviolet, vacuum ultraviolet, and soft x-ray wavelengths. Mixtures of metal vapors and inert gases, and other mixed gas systems, allow generation to regions of the spectrum where nonlinear optical crystals are opaque; and also allow generation at high incident power and energy densities. Progress is reported on programs aimed at efficient conversion from  $1.064\mu$  to  $3547\text{ \AA}$ , and from  $3547\text{ \AA}$  to  $1182\text{ \AA}$ . To date, the shortest wavelength generated by this technique is  $887\text{ \AA}$ . Theoretical considerations indicate that generation into the soft x-ray region should be possible. Using  $1182\text{ \AA}$  radiation, a holographic grating with a fringe spacing of  $836\text{ \AA}$  has been constructed and examined on an electron microscope.

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\* The work reported in this paper was jointly supported by the Office of Naval Research, the U.S. Army Research Office, the Air Force Cambridge Research Laboratories, the Atomic Energy Commission, and the National Aeronautics and Space Administration.

## Second harmonic generation and infrared mixing in $\text{AgGaSe}_2$

R. L. Byer, M. M. Choy, R. L. Herbst, D. S. Chemla\*, and R. S. Feigelson†

*Microwave Laboratory, W. W. Hansen Laboratories of Physics, Stanford University, Stanford, California 94305*  
(Received 17 September 1973; in final form 29 October 1973)

We have continuously tuned between 7 and 15  $\mu\text{m}$  by mixing the output of a  $\text{LiNbO}_3$  parametric oscillator in the chalcopyrite  $\text{AgGaSe}_2$ . We have doubled a  $\text{CO}_2$  laser with 2.7% efficiency which agrees very well with the expected efficiency and verifies the high optical quality of the 1.53-cm-long  $\text{AgGaSe}_2$  crystal. The measured transparency range, indices of refraction, and nonlinear coefficient of  $d_{30} = (3.7 \pm 0.6) \times 10^{-11} \text{ m/V}$  show that  $\text{AgGaSe}_2$  is a useful infrared nonlinear material phase matchable over the entire 3-18- $\mu\text{m}$  infrared region.

EXPERIMENTAL AND THEORETICAL STUDIES OF  
THIRD-HARMONIC GENERATION IN  
THE CHALCOPYRITE  $\text{CdGeAs}_2$

Daniel S. Chemla, R. F. Begley, and Robert L. Byer  
Microwave Laboratory  
Stanford University  
Stanford, California 94305

ABSTRACT

Experimental and theoretical studies of third-harmonic generation (THG) in the chalcopyrite semiconductor  $\text{CdGeAs}_2$  are presented. The phase-matching configurations for THG are analyzed from the irreducible components point of view. A theory of the bound electron and free-carrier contribution to the third-order susceptibility is presented. The experimental results are given. The effective nonlinear coefficient for type-II THG is mainly due to the free-carrier contribution, and for a hole concentration of  $5 \times 10^{16}/\text{cm}^3$  it is measured to be  $(13 \pm 6)10^{-11}$  ESU. The practical applications of THG in  $\text{CdGeAs}_2$  are discussed.

## Resonantly two-photon pumped frequency converter\*

S. E. Harris and D. M. Bloom

*Microwave Laboratory, Stanford University, Stanford, California 94305*  
(Received 26 November 1973)

This letter describes a resonantly two-photon pumped frequency converter with application to the generation of tunable ultraviolet and vacuum ultraviolet radiation and also to infrared-to-visible up-conversion and imaging. Calculations show that up-conversion power efficiencies in excess of 100% should be obtainable with tunable dye lasers having peak powers in the hundred watt to kilowatt range.

## OPTICAL BACKWARD MIXING IN SODIUM NITRITE

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Stanford University, Stanford, California 94305

ABSTRACT :

We present an experimental study of backward mixing of argon laser lines and CO laser lines to generate visible light in sodium nitrite. Angle tuning has been measured. Some special aspects of backward interaction band width are discussed.

A 1.4  $\mu\text{m}$  to 4.4  $\mu\text{m}$  HIGH ENERGY ANGLE TUNED  $\text{LiNbO}_3$   
PARAMETRIC OSCILLATOR

R.L. Herbst, R.N. Fleming and R.L. Byer

ABSTRACT

We have operated a high gain, angle tuned, singly resonant  $\text{LiNbO}_3$  parametric oscillator pumped directly at 1.06  $\mu\text{m}$  by a Q-switched Nd:YAG laser. The oscillator angle tunes from degeneracy and operates over the entire 1.4  $\mu\text{m}$  to 4.4  $\mu\text{m}$  range. Output energies of greater than 1 mJ/pulse at 5 pps have been observed with a 15% energy conversion efficiency. The key to this device is the large  $\text{LiNbO}_3$  crystals fabricated from new [01·4] grown boules. Crystals up to 15 mm diameter and 5 cm in length have been cut at the nominal 47° orientation. These crystals will potentially handle over 2J of optical energy at 1.06  $\mu\text{m}$ .

## GROWTH AND APPLICATION OF [01·4] $\text{LiNbO}_3$

R.L. Byer, R.L. Herbst, R.S. Feigelson and W.L. Kway  
Stanford University  
Stanford, California

### ABSTRACT

We have successfully grown large, high quality  $\text{LiNbO}_3$  boules in the [01·4] direction which lies in the  $yz$  plane  $38^\circ$  to the  $z$  axis. Following annealing and poling the material is strain free, striation free and of high optical quality. It has been used to fabricate an electro-optic switch, for second harmonic generation and tunable parametric generation over the  $1.4 \mu\text{m}$  to  $4.4 \mu\text{m}$  spectral range.

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